

## BEHAVIOR OF BRINES CONTAINING DISSOLVED CO<sub>2</sub> IN ABANDONED WELLBORES

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### ABSTRACT

In order to ensure safe long-term storage of carbon dioxide in geologic formations, the risks posed by improperly abandoned wells must be understood and controlled. Under specific circumstances with formation overpressure or overlying aquifer drawdown, CO<sub>2</sub> that is dissolved in brine can flow up improperly abandoned wells, where it can potentially enter and contaminate USDW. The possibility that depressurization in the wellbore may cause CO<sub>2</sub> exsolution from brine to form a separate buoyant gas phase is of primary concern. Numerical models are used to evaluate these effects in wellbores and to examine the effects of system parameters on CO<sub>2</sub> and brine leakage rates through wellbores.

Up to 20% of the dissolved CO<sub>2</sub> is found to exsolve in simulations of brine transport up a wellbore. The degree of leakage is constrained by the properties of the well, with the permeability of the well being of chief importance. However, at high well permeabilities, the geologic formations provide more resistance to flow than the well and constrain leakage rates. It is found that the presence of stratified permeable layers limit the possibility of upward migration of dissolved CO<sub>2</sub>.

### INTRODUCTION

During carbon capture and storage (CCS), it is expected that injected supercritical CO<sub>2</sub> will eventually dissolve into the storage formation brines over time scales of hundreds to thousands of years (McPherson and Cole, 2000; Ennis-King and Paterson, 2003). Moreover, some researchers have proposed injecting CO<sub>2</sub> as a dissolved phase (Burton and Bryant, 2007; Leonenko and Keith, 2008; Burton and Bryant,

2009). Once CO<sub>2</sub> is dissolved into storage formation brines, a buoyant CO<sub>2</sub> phase no longer exists. Upon dissolution, the brine becomes about 1% denser (Enick and Klara, 1990; Bachu and Adams, 2003); therefore, it will have a tendency to sink slowly to the bottom of the formation.

While it is less likely that dissolved CO<sub>2</sub> will leak up abandoned wellbores, there is still a potential danger. If the storage formation is overpressured, or if an overlying aquifer is drawn down due to pumping, the pressure differential could induce the brine containing the dissolved CO<sub>2</sub> to flow upward through permeable pathways such as abandoned wells. As pressure decreases, and the leaked brine moves upward, CO<sub>2</sub> solubility decreases, causing gas exsolution (Pruess, 2008). This evolved gas phase has the potential to accumulate in underground sources of drinking water (USDW) or potentially migrate to the surface due to its buoyancy.

The focus of this research is to examine the properties in an abandoned wellbore that control the overall flow rate for leakage of brine containing dissolved CO<sub>2</sub>. In addition, gaseous CO<sub>2</sub> exsolution effects and leakage plumes are evaluated. This work builds on studies such as that by Birkholzer et al. (2011), whose focus is brine leakage through wellbores near the area of review boundary. Unlike that study, the present work is specifically directed toward wellbore leakage of CO<sub>2</sub>-laden brine. Investigation of how this dissolved-phase CO<sub>2</sub> could leak up wellbores and contaminate overlying USDW is of primary concern. Wellbore leakage of dissolved CO<sub>2</sub>-laden brine due not only to stor-

age formation overpressure, but also to overlying aquifer drawdown, are examined.

In the present study, changes in flow due to system parameters are examined in detail. Flow effects when multiple permeable formations are present along the wellbore are considered.

### **DIRECT LEAKAGE MODEL FROM STORAGE FORMATION TO USDW**

In order to perform a detailed analysis of CO<sub>2</sub>-laden brine leakage into USDW through abandoned wells, we must consider effects that cannot be accounted for in analytical solutions. Important flow effects may be caused by pressure and temperature gradients, phase changes, salinity, depressurization, and multiphase flow phenomena.

Our numerical model is constructed using the TOUGH2-ECO2N multiphase flow simulator (Pruess, 2007) with the PetraSim GUI (Swenson, D. 2003). The TOUGH2-ECO2N simulator allows for modeling the development of dissolved and gaseous CO<sub>2</sub> plumes in the USDW due to overpressurization of the storage formation or USDW drawdown.

A radially symmetric numerical grid design is used for the simulations, which is similar to numerical grids used in other studies (Birkholzer et al., 2011). In Figure 1, the model design can be seen. This model represents an endpoint case in which a dissolved CO<sub>2</sub> storage formation is directly connected to a USDW through a wellbore. This allows for an evaluation of flow effects with simple, but representative geometry.

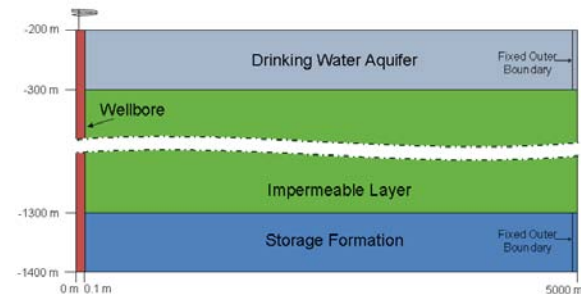


Figure 1. Base-case numerical model setup. The storage formation and USDW are separated by a 1000 m thick impermeable layer. All formations are penetrated by a well in the central radial element.

The outer radius of the model is set to 5,000 m. This radius is sufficiently large to minimize numerical end effects. The model is given an overall vertical thickness of 1,200 m. Both the storage formation and the drinking water aquifer are 100 m thick. Between the two formations is a 1000 m thick impermeable layer. The impermeable layer between the formations represents a case in which only flow between the storage formation and the USDW are considered. The layer does not transmit fluids, but it does allow for thermal conduction of heat from the warmer storage formation brine as it rises through the wellbore. Finally, the top of the model is set at a depth of 200 m below the ground surface, which is a representative depth for a large regional USDW. Hydrostatic pressure is used to generate the pressure gradient, such that the pressure at the top of the upper formation is  $2.0 \times 10^6$  Pa, and at the bottom of the lower formation it is  $1.4 \times 10^7$  Pa. A geothermal gradient of 30°C/km with a surface temperature of 15°C is used. This gradient is typical of the western United States and has been used in other numerical models (Pruess, 2008). Finally, CO<sub>2</sub> exists just below its solubility in the storage formation in order to simulate CO<sub>2</sub>-laden brine.

The wellbore is modeled as the central radial element in the model, and it is given a different permeability from the surrounding elements. As in previous studies (Nordbotten et al., 2005; Ebigo et al., 2007; Nordbotten et al., 2009; Birkholzer et al., 2011), it is assumed that Darcy's law applies to flow in the well.

However, when well permeabilities above  $1 \times 10^{-7} \text{ m}^2$  are used in the numerical simulations, it is assumed that they are representative of open pipe flow. We acknowledge that the numerical assumption of laminar flow according to Darcy's law in the wellbore may not fully capture flow effects for the case of turbulent flow or for different multiphase flow regimes in an open wellbore.

The model is discretized by using 87 elements in the radial dimension. The radial gridblocks are refined around the well with each successive

ring increasing in thickness to maximum of 95 m. The outermost ring is used to impose a fixed state boundary on the model. The fixed state conditions (constant pressure, temperature, salt concentration, CO<sub>2</sub> saturation) in this ring allow for flow into the storage formation as well as flow out of the USDW. Similar to Birkholzer et al. (2011), the pressure in the outer ring can be increased to overpressurize the storage formation or lowered to induce drawdown in the USDW.

In the vertical z dimension, 70 elements are used. The storage formation uses ten 10 m thick model layers, whereas the USDW is refined using twenty 5 m thick model layers. The impermeable layer consists of forty 25 m thick model layers (these mainly conduct heat from the wellbore). This discretization results in a total of 6090 gridblocks for the entire model.

Relative permeabilities and capillary pressures for the CO<sub>2</sub> and brine are calculated using modified versions of the van Genuchten equations, and are the same as those used in simulations in Doughty, (2007). Capillary pressure is zero in the well, and, the well porosity is set to 0.98. Other relevant model properties can be seen in Table 1.

Table 1. Material properties

Property	Value
<b>Entire Model</b>	
Thermal Conductivity (W/m·°C)	2.51
Heat Capacity (J/Kg·°C)	920
Rock Density (Kg/m <sup>3</sup> )	2600
<b>Drinking Water Aquifer</b>	
Permeability (m <sup>2</sup> )	1x10 <sup>-12</sup>
Porosity	0.25
Salinity (mg/l)	0
<b>Impermeable Layer</b>	
Permeability (m <sup>2</sup> )	1x10 <sup>-20</sup>
Porosity	1x10 <sup>-4</sup>
<b>Storage Formation</b>	
Permeability (m <sup>2</sup> )	1x10 <sup>-13</sup>
Porosity	0.25
Salinity (mg/l)	20,000
CO <sub>2</sub> Mass Fraction	0.044
<b>Wellbore</b>	
Permeability, (m <sup>2</sup> )	1x10 <sup>-5</sup> – 1x10 <sup>-12</sup>
Porosity	0.98
Well Diameter (m)	0.2 (8 inch)

Simulations can be run with different values of the well's permeability over a range of system overpressures and drawdowns ranging between 10 and 30 bar.

#### Wellbore Permeability Effects on Leakage

Knowing the apparent permeability of an improperly abandoned well is difficult, since so little is known about the spatial distribution and properties of these wells (Ide et al., 2006). Other researchers have dealt with leaky wellbore permeability by using a range of permeability values (Celia et al., 2004; Birkholzer et al., 2011), and that approach is used here.

This numerical model uses an 8-inch (0.2 m) diameter well, and the storage formation is overpressurized 20 bar to induce flow up the wellbore. This overpressure is consistent with predicted long-term overpressures seen in other studies (Zhou et al., 2010).

In the wellbore, a significant fraction of the CO<sub>2</sub> exsolves to form a separate supercritical or gas phase as the brine is depressurized during upward migration due to storage formation overpressure. For leakage simulations at high well permeabilities, the gas fraction that accumulates in the USDW is higher than for leakage with lower well permeability. Higher gas fractions at high well permeabilities are due to a larger increase in temperature in and around the wellbore. Because flow rates are largest at high permeabilities, more warm brine leaks, which heats up the system and reduces CO<sub>2</sub> solubility.

However, re-dissolution of the exsolving CO<sub>2</sub> as it contacts resident USDW fluid during leakage reduces the gas phase in the USDW. At lower well permeabilities, and thus flow rates, this effect becomes dominant. When the flow rates are very low due to low well permeability, all of the exsolving gas becomes redissolved in the aquifer during leakage (Figure 2).

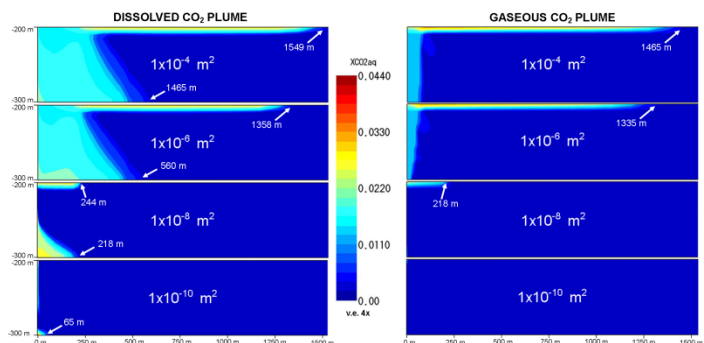


Figure 2. Dissolved and gaseous CO<sub>2</sub> plume in the drinking water aquifer at various well permeabilities at 50 years. Plume formation is not significant at well permeabilities below  $1 \times 10^{-10} \text{ m}^2$ .

After 50 years of wellbore leakage into the USDW, CO<sub>2</sub> leakage plumes have significantly different magnitudes for cases of different wellbore permeabilities. For well permeabilities below  $1 \times 10^{-8} \text{ m}^2$ , the CO<sub>2</sub> plume mass decreases linearly with decreasing well permeability. However, in simulations with well permeabilities above  $1 \times 10^{-7} \text{ m}^2$ , leakage plumes do not continue to increase in magnitude significantly. In addition, examination of leakage velocities into the USDW show a linear increase in velocity with increasing permeability up to  $1 \times 10^{-7} \text{ m}^2$ , suggesting that the permeability of the wellbore controls the leakage rate. However, at high well permeabilities, the leakage rate remains nearly constant, suggesting that the permeability of the well no longer controls the flow, but rather the permeability of the geologic formations is the limiting factor.

The USDW leakage plumes tend to have two separate regions. Along the top of the USDW, a gaseous plume occurs which increases in size with increasing well permeability. Secondly, there is a wedge shaped dissolved plume that extends downward to the base of the USDW. It also increases with increasing well permeability.

The exsolved gaseous plume spreads along the top of the aquifer due to being highly upwardly buoyant compared to the surrounding water. The wedge-shaped dissolved plume migrates downward in the USDW because the CO<sub>2</sub>-laden brine

that is leaking has a higher density than surrounding waters.

The total CO<sub>2</sub> mass flow rate (dissolved and gaseous) in the well at the base of the USDW over 50 years for well permeabilities ranging between  $1 \times 10^{-4}$  to  $1 \times 10^{-12} \text{ m}^2$  show that at higher well permeabilities ( $1 \times 10^{-4}$  to  $1 \times 10^{-6} \text{ m}^2$ ), the flow rates are fully established within 3 months. However, as well permeability decreases, the time scale for fully established flow is increased indicating that it takes longer for leaked brine to flow upward at lower permeabilities.

At well permeabilities above  $1 \times 10^{-7} \text{ m}^2$ , the flow rate never reaches steady state but instead exhibits small oscillations as well as a gradual decline over time. This is due to thermal effects similar to those shown by Oldenburg and Rinaldi, (2010), where warm brine cools upon entry into the USDW and moves downward due to an increase in density. Additional cooling occurs due to gas exsolution. As wellbore permeabilities below  $1 \times 10^{-7} \text{ m}^2$ , leakage rates of CO<sub>2</sub> reach a steady state once fully established.

Despite these limitations, it is reasonable to surmise that at open wellbore conditions, the well will provide far less resistance to flow than the geologic formations, indicating minimal control over the system. This suggest that even accounting for resistance due to turbulent flow, the geologic formations will provide an upper limit to the leakage rates of CO<sub>2</sub> laden brine.

In order to evaluate the mass of CO<sub>2</sub> that leaks into the USDW as a function of well permeability and storage formation overpressure, many leakage simulations are performed over a matrix of pressure and well permeability values (Figure 3).

After 25 years, the mass of leaked CO<sub>2</sub> ranges from zero at low permeabilities and overpressures to as much as 0.5 Mt for permeabilities meant to represent leakage through an open wellbore, with large formation overpressure.

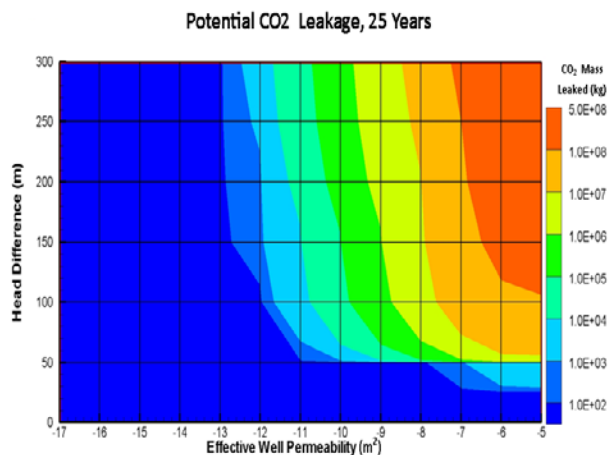


Figure 3. Simulation results showing the simulated mass of CO<sub>2</sub> leakage plumes in the DWA as a function of well permeability and formation overpressure.

Although a large mass of CO<sub>2</sub> leaks for the open-well cases, this represents a worst-case scenario. Leaky well permeability ranges that are probably more realistic for abandoned wells, such as  $1 \times 10^{-10}$  to  $1 \times 10^{-14}$  m<sup>2</sup>, have been used in other studies of wellbore leakage (Celia, M.A. 2004; Nordbotten, J.M. 2005; Nordbotten, J.M. 2009). For this permeability range, the mass of CO<sub>2</sub> leaked is at least three orders of magnitude lower than for open wellbore flow leakage. Therefore, leaky wells with some degree of blockage likely pose a far lower leakage risk than for leakage through an open wellbore.

### Leakage Plume Behavior Post Injection

Once overpressure or drawdown has been established such that CO<sub>2</sub>-laden brine is flowing in a wellbore, there is a concern that as CO<sub>2</sub> exsolves, it might generate a self-enhancing system whereby exsolution causes a gas drive that persists beyond the overpressure or drawdown event. In order to examine these effects as well as leakage plume behavior, the model is overpressured by 20 bar and the simulation is run for 100 years of overpressure.

The dissolved CO<sub>2</sub> leakage plume in the USDW extends out 315 m after 100 years of leakage. In addition, gaseous CO<sub>2</sub> has developed and accumulated along the top of the USDW with a radial extent of 300 m. At the end of the 100-

year injection, the outer radial ring pressure in the storage formation is returned to hydrostatic conditions. Within 60 days, the reduction in overpressure reaches the wellbore. As soon as this occurs, the upward flow of CO<sub>2</sub> laden brine ceases.

No solution-gas-drive effects are observed in the simulated system. Because the dissolved CO<sub>2</sub> in the storage formation is only a small fraction of the total fluid mass (4.4%), with less than 20% of that coming out of solution in the wellbore and USDW, the overall flow rate is dominated by the brine and is not controlled by exsolving CO<sub>2</sub>. Therefore, for the simulated scenario, in the absence of an external pressure disturbance, the exsolving CO<sub>2</sub> is unable to establish upward gas drive.

Once overpressure ceases, water and CO<sub>2</sub> in the USDW begin flow back down the wellbore and eventually reach a steady downward flow rate of 0.19 kg/s (~3 gal/min) for the water. Due to CO<sub>2</sub> flow back down the wellbore, after 25 years, 7% of the total leaked CO<sub>2</sub> has been flushed down the wellbore while 15.8% has been flushed at 100 years. However, the gaseous plume is diminished much faster than the dissolved CO<sub>2</sub> plume. After 25 years, 29% has left the USDW while 54.5% is removed after 100 years (Figure 4).

Initially, when the overpressure is removed, the leaked CO<sub>2</sub>-laden brine in the USDW flows down the well due to its density and the lack of a continued pressure head for inducing upward flow. However, once this initial effect ceases, the fluid around the wellbore in the USDW remains denser than the fluid below it due to its lower temperature, allowing for downward migration. As water in the USDW contacts the gaseous CO<sub>2</sub> plume, the CO<sub>2</sub> is stripped into the dissolved phase in the water, increasing its density. This fluid then drains down the wellbore, allowing more unsaturated water to contact the gaseous plume. Therefore, the bottom of the gaseous CO<sub>2</sub> plume along the top of the aquifer is constantly being contacted by unsaturated water moving past it. The reduction in the gase-

ous and dissolved CO<sub>2</sub> plume in the USDW can be seen in Figure 4.

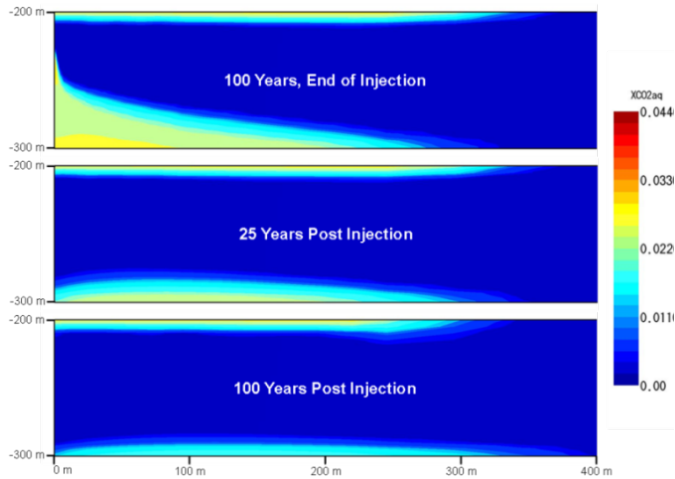


Figure 4. Dissolved CO<sub>2</sub> plume migration in the entire system and in the USDW after 100 years of leakage due to overpressure.

In addition to reducing the plume in the USDW, the area around the wellbore in the dissolved CO<sub>2</sub> storage formation is being diluted with respect to CO<sub>2</sub> content. This is due to the downward migration of water from the USDW. For this simulated case, the downward migration of fluid post injection further reduces the leakage risks posed by dissolved CO<sub>2</sub>. However, we note that this backflow phenomena may not occur if the storage formation brine has a high density, due to a higher salinity.

#### **STRATIFIED WELLBORE LEAKAGE MODEL**

For this case, the region containing the impermeable layer is modified so that effects due to interbedded stratigraphy can be examined. A formation with the same properties as the USDW is assigned to four 100 m thick zones along the height of the model with top elevations of 350, 550, 750, and 950 m below the ground surface respectively. All formations are penetrated by the 8-inch diameter  $1 \times 10^{-8}$  m<sup>2</sup> permeability well, and the wellbore is open to each formation.

#### **Overpressure with Dissolved CO<sub>2</sub>**

The stratified model is run for fifty years with an overpressure of 20 bar. After 50 years, no CO<sub>2</sub>, dissolved or gaseous, has reached the drinking water aquifer. The four interbedded aquifers provide significant leakage for the CO<sub>2</sub> laden brine as it moves up the wellbore. Because the majority of the CO<sub>2</sub> is dissolved in the leaking brine, the effects of capillary entry pressure and relative permeability are minimal. Therefore, the majority of the dissolved CO<sub>2</sub> is transported into interbedded layers as a part of the single aqueous phase brine. Furthermore, the largest amount of leaked brine migrates directly into the closest overlying formation. Some gaseous CO<sub>2</sub> leaks into the stratified layers as well. CO<sub>2</sub> exsolution occurs within the wellbore during upward migration. However, instead of continuing to migrate upward, the gaseous CO<sub>2</sub> enters into the interbedded stratigraphy. Both the dissolved and gaseous CO<sub>2</sub> plumes in the stratified system can be seen in Figure 5.

In the system modeled, the presence of interbedded stratigraphy reduces the leakage risk for CO<sub>2</sub> into a USDW. Further simulations are run with overpressures as high as 50 bar. Even at this high overpressure, the CO<sub>2</sub> never moves into the drinking water aquifer, but instead has a greater degree of accumulation in the interbedded aquifers between the storage formation and the USDW.

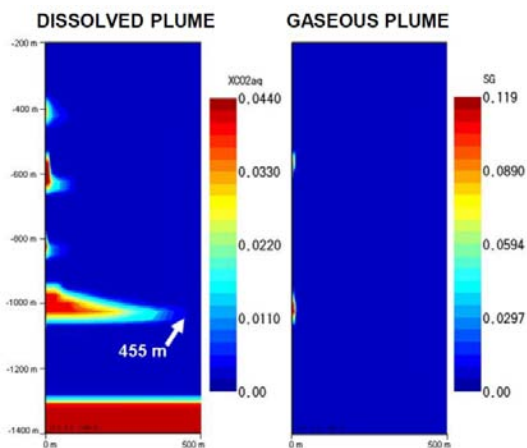


Figure 5. Dissolved and gaseous leakage plumes at 50 years for the stratified simulation at 50 years of leakage.



### Effects of USDW Drawdown

The stratified model is used to simulate leakage effects due to USDW drawdown when permeable stratified layers are present. In order to examine a worst-case scenario, the wellbore is made highly permeable ( $1 \times 10^{-5} \text{ m}^2$ ) to maximize the possible amount of upward flow due to drawdown. The USDW is then drawn down 20 bar (~200 m head).

Simulations show that as fluid flows into the upper aquifer, it is being pulled directly from the interbedded aquifers below. In this scenario, only a small amount of CO<sub>2</sub>-laden brine is drawn out of the storage formation, but it is never pulled up the well far enough to enter overlying permeable layers. Upward flow of fluid from each formation is reduced with depth as each successive permeable layer below the USDW contributes less to overall flow rate. Whenever interbedded permeable layers exist between a formation being drawn down and a dissolved CO<sub>2</sub> storage formation, the possibility of dissolved CO<sub>2</sub> leakage is greatly reduced by the interbedded permeable layers.

### SUMMARY

Simulations have been performed to evaluate leakage of CO<sub>2</sub>-laden brine through poorly sealed or improperly abandoned wellbores. Through numerical models, it has been found that the overriding controls of wellbore leakage of brines containing dissolved CO<sub>2</sub> are storage formation overpressure and well permeability

In most leakage scenarios where the wellbore is not an open wellbore but instead is blocked in some fashion, well permeability controls the leakage rate of CO<sub>2</sub>-laden brine into permeable formations. However, if the endpoint case of leakage through a completely open well occurs, the permeabilities of the geologic formations will provide more resistance to fluid flow. As a result, there is a probable upper limit to the leakage flow of brine, even with an open well.

For the endpoint case where no permeable layers are present between the storage formation and the USDW, significant amounts of gas can

exsolve during leakage to form a separate gas phase in the USDW. However, in a more realistic case where interbedded permeable layers are present, simulation results show that while gas phase exsolution does occur, large gaseous plumes are not observed in the USDW.

In the simulations performed, CO<sub>2</sub>-laden brine leakage due to drawdown of overlying USDW poses minimal risks if stratified permeable layers are present. During drawdown, fluid is preferentially drawn out of formations directly underneath the formation being pumped. Thus, in a typical stratified system, it is possible that a deep CO<sub>2</sub> storage formation will not be affected by drawdown in an overlying USDW.

After overpressure has ceased, leakage of CO<sub>2</sub> laden brine does not continue. No solution gas drive effects are observed in the simulations. Furthermore, after injection has ceased, significant amounts of the leaked CO<sub>2</sub>, especially the gaseous plume, may be flushed back down the wellbore due to a depth decreasing density gradient in the system. This serves as a natural mechanism for CO<sub>2</sub> leakage mitigation.

Numerical simulations are unable to capture the exact behavior of dissolved CO<sub>2</sub> leakage for an open wellbore. Although effects due to turbulent friction as well as multiphase flow regimes are not considered, the high-permeability model results still provide valuable insights into the behavior of dissolved CO<sub>2</sub> leakage.

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